



LAWRENCE
LIVERMORE
NATIONAL
LABORATORY

Methodology and Determination of Field of View of Neutron and Gamma Detectors in the Atucha Spent Fuel Storage Pool

W. Walters, M. Wenner, A. Haghighat, S. Sitaraman, Y. S. Ham

June 29, 2009

Methodology and Determination of Field of View of Neutron and Gamma Detectors in the Atucha Spent Fuel Storage Pool
Tucson, AZ, United States
July 12, 2009 through July 16, 2009

Disclaimer

This document was prepared as an account of work sponsored by an agency of the United States government. Neither the United States government nor Lawrence Livermore National Security, LLC, nor any of their employees makes any warranty, expressed or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States government or Lawrence Livermore National Security, LLC. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States government or Lawrence Livermore National Security, LLC, and shall not be used for advertising or product endorsement purposes.

Methodology and Determination of Field of View of Neutron and Gamma Detectors in the Atucha Spent Fuel Storage Pool

William Walters, Michael Wenner and Alireza Haghighat
Department of Nuclear and Radiological Engineering, University of Florida

Shivakumar Sitaraman and Young Ham
Lawrence Livermore National Laboratory

For the Institute of Nuclear Materials Management (INMM) 50th Annual Meeting
Major Topical Area: Modeling and Analysis
Sub-Topic: Materials Control and Accountability – Measurements and Instrumentation

Abstract

In this paper we seek to create a model by determining the field of view (FOV) of a detector (i.e. which assemblies contribute to the detector response) in the Atucha-I spent fuel pool. The FOV is determined by solving the adjoint transport equation using the 3-D, parallel PENTRAN (Parallel Environment Neutral-particle TRANsport) Sn code, with the detector cross section as the adjoint source. If this adjoint function is coupled with the source spectrum, then the contribution to the detector from each assembly can be determined. First, the reactor criticality was modeled using the MCNP5 (Monte Carlo N-Particle) Monte Carlo code in order to determine the power distribution in each assembly. Using the power distribution data, the assemblies were divided and homogenized into 8 axial and 3 radial zones for burnup analysis. Depletion calculations were performed for each zone using the ORIGEN-ARP (Automatic Rapid Processing) utility from the SCALE 5.1 (Standardized Computer Analyses for Licensing Evaluation) code package. Spent fuel pool and detector were modeled in 2-D in PENTRAN as the detector plus 3 fuel assemblies along both x and y axes. Using the resulting adjoint function combined with the source spectrum, we have determined the FOVs of the fission chamber neutron detector that was used at Atucha, and concluded that 2 assemblies along x and y axes are needed for both cases (i.e. the 4 adjacent assemblies plus the next surrounding 12). For the neutron detector, 88% of the response comes from the nearest 4 assemblies, with 99% from the nearest 16). Results for a uniformly sensitive gamma detector indicate that 2 assemblies in both directions are also needed, with 89% of the response coming from the adjacent assemblies. A Monte Carlo calculation using MCNP was performed to benchmark the neutron result, giving a similar result (87% MCNP vs. 88% PENTRAN). Based on these studies, we have developed a database of FOVs as a function of burnup and decay conditions for different detector types, and a methodology/algorithm which uses this database to analyze the response of a detector placed in a spent fuel pool with the aim of detecting gross defects.

Introduction

Methods for spent fuel verification can be difficult for reactors that have a spent fuel storage arrangement that does not permit easy movement and isolation of assemblies, such as at the Atucha-I reactor in Argentina. Spent fuel verification is extremely

important with regards to proliferation, in order to make sure nuclear material is not being diverted for illicit use. Previous work by Ham et al.[1] used a neutron detector placed in the assembly lattice, and assumed that the count rate would be proportional to the sums of the burnups of the 4 surrounding assemblies. While this approach is effective for low burnup fuel (5-8 GWd/t), it is not valid for higher burnups. This study seeks to create a more general methodology that is valid for all burnup levels and cooling times found in the spent fuel pool at Atucha-I.

Spent Fuel Characterization

Fuel assemblies at Atucha-I are circular with 36 fuel elements and one support rod. The 5.3m long assemblies are arranged in a triangular lattice of pitch 27.2cm with heavy water acting as coolant and moderator. Fuel pellets are UO₂, either natural uranium (NU) or slightly enriched uranium (SEU, 0.85% enriched).

In order to determine the burnup distribution of the fuel, an MCNP model was made with one fuel assembly in an infinite lattice, as shown in Figures 1 and 2.

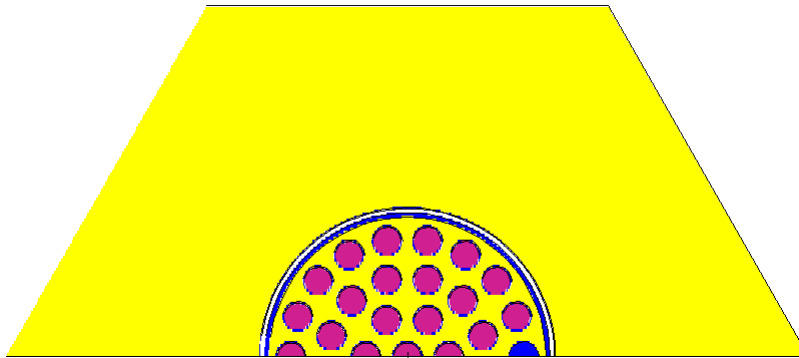


Figure 1: x-y view of MCNP criticality model. Fuel is in purple and heavy water is yellow. Top/bottom/sides are all reflected.

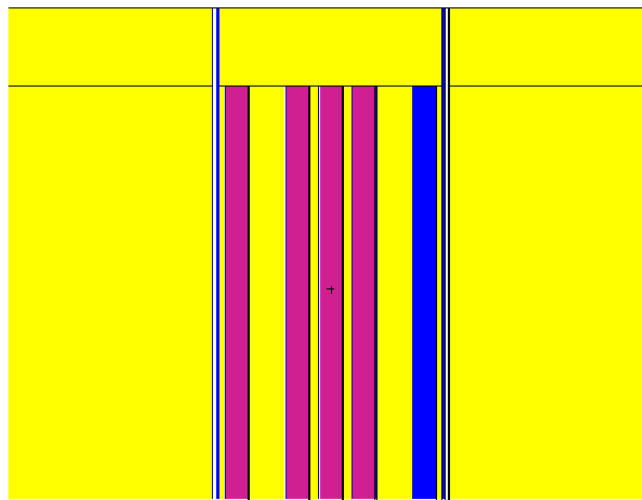


Figure 2: x-z view of MCNP criticality model. Fuel is in purple and heavy water is yellow. Bottom/sides are all reflected. The top boundary is vacuum.

Using fresh fuel, the fission rate was calculated throughout the assembly. This fission distribution was assumed to not change throughout the burn cycle and to not be effected by control rods. The fission distribution by fuel pin and axial position is shown in Figure 3. Fuel pins closest to the structural pin had the highest fission rate, due to a lack of absorption that would otherwise occur at the pin location. However, the biggest difference was the radial position, with the most fission occurring in the outer fuel pins. For the model, the difference in burnup of pins at a given radial position was ignored.

In order to simplify the problem, each assembly was homogenized into 3 radial rings and 8 axial levels, as shown in Figure 3. A simple volume homogenization was performed.

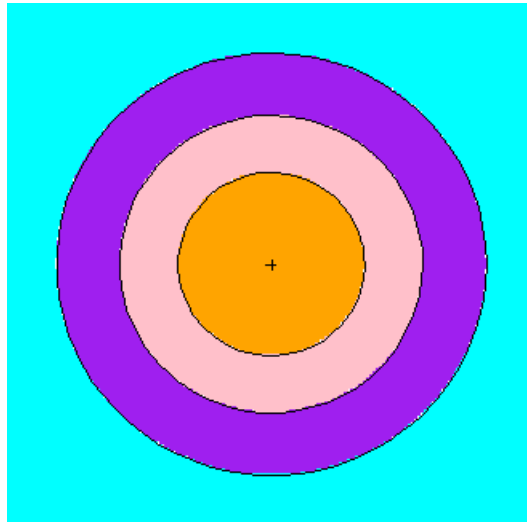


Figure 3: Homogenized version of a fuel assembly.

Using the fission distribution and the total known burnup of the assembly, it was possible to assign a specific local burnup value to each zone (3 radial zones and 8 axial levels). For each of these zones, a burnup and decay calculation was performed using ORIGEN-ARP (Automatic Rapid Processing). ORIGEN-ARP has cross section libraries of standard fuel assemblies across a range of burnups. The Atucha-I type fuel was not available, but the CANDU 37-element design was, which is very similar to Atucha-I. These calculations produced a database of material compositions and source strength as a function of location within the assembly, total burnup and decay time. Figure 4 shows the total neutron source strength as a function of local burnup and decay time. Figure 5 shows the gamma source. Both figures are for natural uranium fuel.

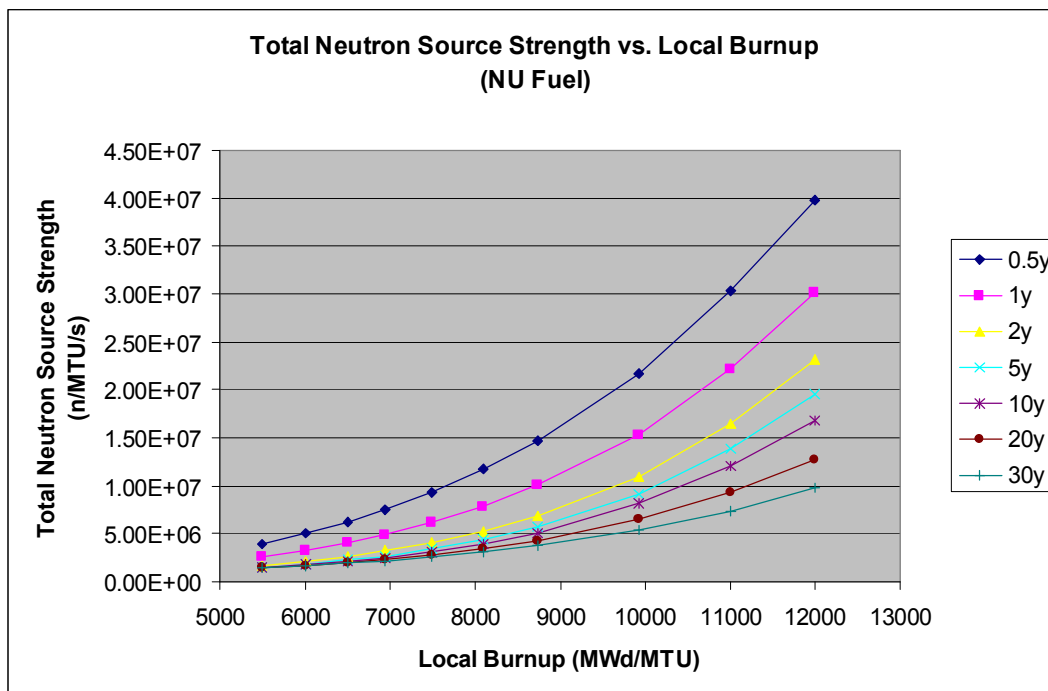


Figure 4: Total neutron source strength as a function of decay time and local burnup for NU fuel

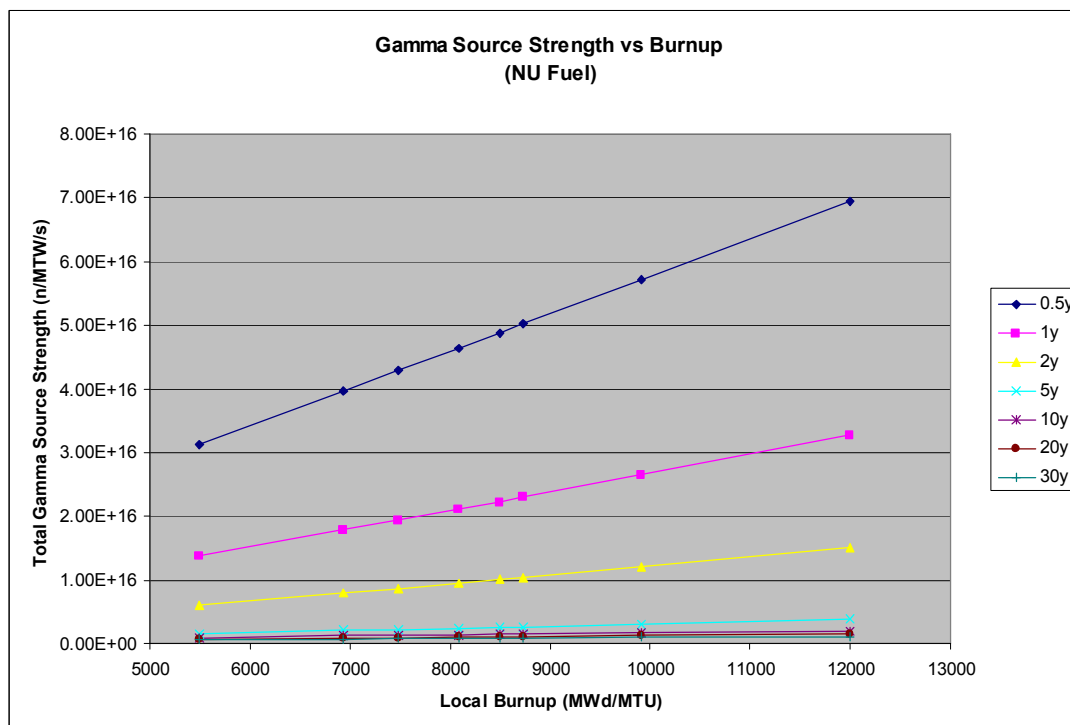


Figure 5: Total gamma source strength as a function of decay time and local burnup for NU fuel

It can be seen that the gamma source is approximately linear with burnup and decays very quickly. The neutron source is non-linear (slope increasing with burnup) and decays slower than the gamma source. In terms of relative magnitude, the gamma source is much larger, by a factor of approximately 10^9 .

Cross sections for the problem were taken from the BUGLE-96 library, with 47 neutron groups and 20 photon groups. The GMIX utility was used to mix the cross sections of materials calculated with ORIGEN-ARP. Cooling time for the materials was assumed to be 10 years. It will be shown later that both cooling time and burnup does not have a large effect on the cross sections.

Detector Importance Calculations

A standard forward calculation of detector response would first involve solving for the angular flux Ψ given a source S . Dependencies on (r, Ω, E) are implicit and left out for brevity.

$$H\Psi = S \quad (1)$$

Where,

$$H\Psi = [\Omega \cdot \nabla + \sigma(r, E)]\Psi(r, \Omega, E) - \int dE' \int d\Omega' \sigma_s(r, E' \rightarrow E, \Omega' \cdot \Omega) \Psi(r, \Omega', E') \quad (2)$$

After Ψ has been calculated, the detector response can be calculated as:

$$R = \langle \Psi \sigma_d \rangle \quad (3)$$

Where, $\langle \rangle$ denotes integration over all independent variables (space, energy and angle) and σ_d is the detector cross section.

In the adjoint or importance function methodology, we calculate the detector response by first determining the importance of neutrons to the detector Ψ^* .

$$H^*\Psi^* = S^* \quad (4)$$

H^* is defined by the identity:

$$\langle \Psi^* H\Psi \rangle = \langle \Psi H^*\Psi^* \rangle \quad (5)$$

This yields the adjoint or importance transport operator:

$$H^*\Psi^* = [-\Omega \cdot \nabla + \sigma(r, E)]\Psi^*(r, \Omega, E) - \int dE' \int d\Omega' \sigma_s(r, E \rightarrow E', \Omega' \cdot \Omega) \Psi^*(r, \Omega', E') \quad (6)$$

If we define $S^* = \sigma_d$ then we can obtain the detector response:

$$R = \langle \Psi^* S \rangle \quad (7)$$

Where, S is the standard forward source.

So, $\Psi^*(r, \Omega, E)$ can be taken to mean the probability of a neutron born at phase space (r, Ω, E) within $(dr, d\Omega, dE)$ of causing a reaction in the detector.

The importance function methodology allows for rapid calculations of response given a variety of possible sources. This is very important for fuel verification since the source is not known completely. Using the importance function we can determine the proportion of the detector response that comes from each assembly (i.e. the detector field of view). The fraction of the response due to one particular assembly is:

$$FR_i = \frac{\langle \Psi^* S_i \rangle}{\sum_i \langle \Psi^* S_i \rangle}$$

Where, S_i is the source located in assembly i .

A 2D model was created using PENTRAN to evaluate the detector importance. The detector was a 2.54 cm diameter fission chamber surrounded by a 6 cm diameter polyethylene moderator. Two spacing arrangements were tested: 145 mm in the x direction and 150 mm in the y direction for the NU fuel, and 135 mm in both directions for the SEU fuel. The PENTRAN model of the SEU arrangement is shown in Figure 6 below.

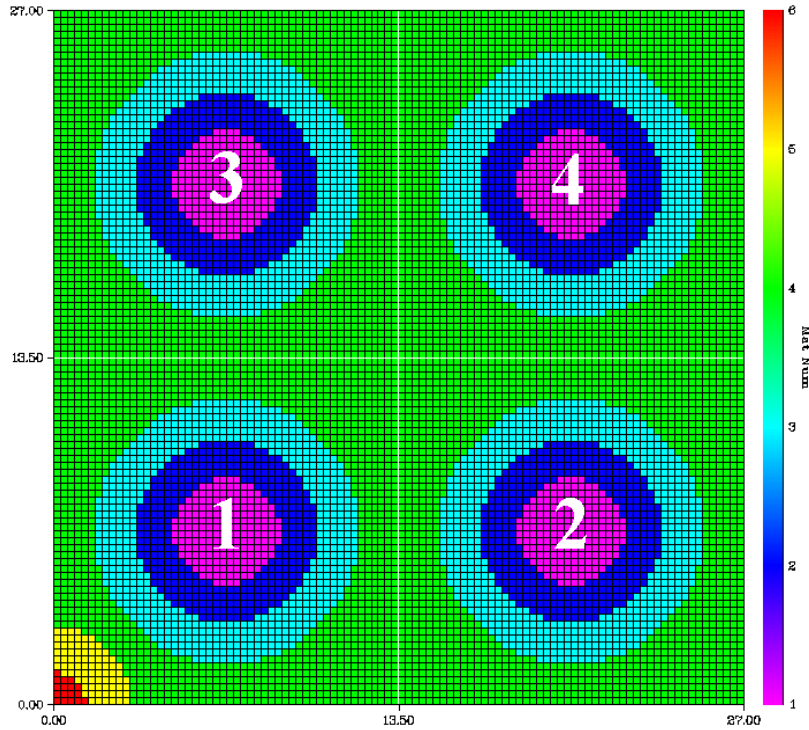


Figure 6: 2D model of the SEU spent fuel pool in PENTRAN. The detector is in red at the bottom left corner. Assembly numbers are also shown.

In order for the calculation to yield to proper detector importance, the source must be equal to the cross section of the detector. In this case, the detector is a fission chamber using 94 w% enriched U. So, the fission cross section of 94% enriched U was used as the adjoint source.

In order to test for proper convergence, several calculations with different mesh sizes and quadrature orders must be tested. This was performed for a mesh size of 0.135cm and 0.270cm; angular quadrature sets tested were S4 and S8. For these cases, the fractional response of each detector was calculated, assuming identical sources in all assemblies. Results are shown in Table 1.

Table 1: Fractional Response of Assemblies for Varying Mesh Size and Quadrature (SEU Fuel)

Fine Mesh Size (cm)	Quadrature Order	Assembly #			
		1	2	3	4
0.270	S4	86.55%	6.13%	6.13%	1.19%
0.270	S8	86.74%	5.99%	5.99%	1.27%
0.135	S4	86.56%	6.13%	6.13%	1.19%
0.135	S8	86.75%	5.99%	5.99%	1.27%

These results show almost identical numbers, indicating that a fine mesh size of 0.270cm and S4 quadrature should be adequate.

Next, calculations were performed to check variations on the model. First, a model with 3x3 assemblies instead of 2x2 assemblies was created, to see how much response was “missing” outside of the nearest assemblies. Another calculation was performed using fresh fuel in order to see the effect of variation of burnup and/or cooling time on the FOV. Results are shown in Table 2.

Table 2: Fractional Response for Different Model Size and Cross Sections

Assembly Arrangement	Assembly #				
	1	2	3	4	All Others
2x2	86.74%	5.99%	5.99%	1.27%	
3x3	85.72%	5.94%	5.94%	1.21%	1.18%
2x2 (Fresh Fuel)	85.63%	6.52%	6.52%	1.33%	

Expanding the model from 2x2 to 3x3 resulted in only ~1% increased response. This was deemed to be insignificant for our purposes, given detector error etc. The fresh fuel numbers are also very close, especially given the very large difference (0 MWd/MTU vs 11000 MWd/MTU). This indicates that any differences in assembly can be ignored in terms of cross sections, especially in the relatively small range of burnups that will be encountered (9000-13000 MWd/MTU).

In order to validate the 2D model, a 3D model was created to make sure that there was not a large effect. The 3D model is shown in Figure 7 below. The active portion of the detector is in red, the inactive portion is in pink, the polyethylene is in orange and a tungsten shield (to shield electronics) is in white. Top, bottom, +x and +y directions had vacuum boundaries while -x, -y had reflective conditions.

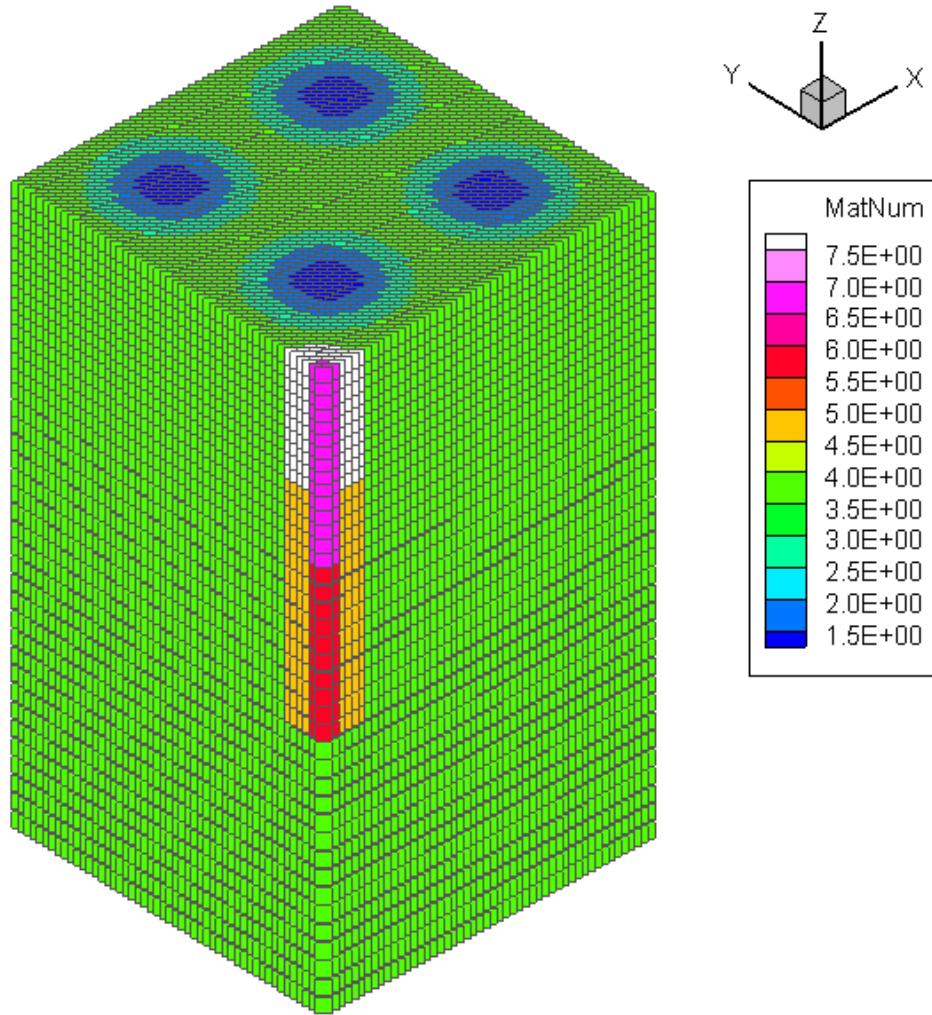


Figure 7: 3D model of detector and spent fuel in PENTRAN

Results from this calculation are show in Table 3. The field of view is almost identical, indicating that a 2D model is adequate while requiring much less computational effort.

Table 3: Field of view for 2D and 3D models (SEU Fuel)

Assembly Number	FR _i (3D)	FR _i (2D)
1	86.75%	86.74%
2	6.05%	5.99%
3	6.05%	5.99%
4	1.14%	1.27%

If the spectrum of neutrons changed significantly with burnup or cooling time, then the resulting FOV could change due to a different source term. To check this, the importance was coupled with sources with different cooling times and burnups. The results, shown in Table 4, indicate that there is very little spectral difference for changing cooling times and burnups. Results are for the NU fuel arrangement (hence the difference between assemblies 2 and 3 due to different pitches in each direction).

Table 4: FOV Variation with Fuel Burnup and Cooling Time
(NU Fuel)

Average Burnup (MWd/MTU)	Cooling Time (years)	Assembly			
		1	2	3	4
5000	10	88.65%	5.31%	5.15%	0.90%
8000	10	88.66%	5.29%	5.14%	0.92%
6000	1	88.54%	5.34%	5.19%	0.93%
6000	30	88.47%	5.39%	5.23%	0.91%

In order to benchmark these importance function results, a forward Monte Carlo transport calculation was performed using MCNP. The model used can be seen in Figure 8. The yellow and blue circle is the detector. The numbered assemblies represent the source location for each of 4 calculations.

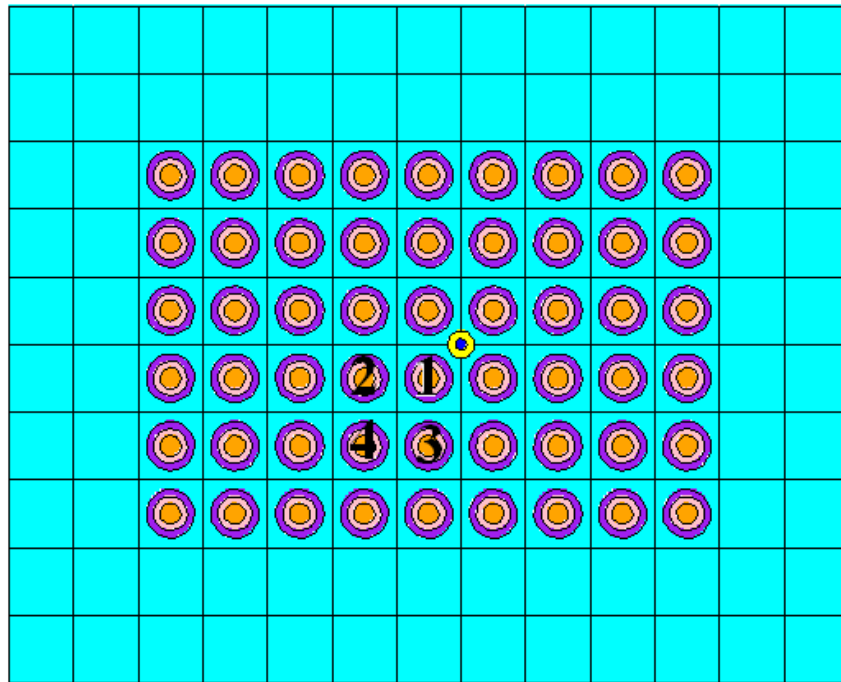


Figure 8: MCNP spent fuel and detector model. Detector is shown in blue and yellow.

Flux was tallied in the detector and multiplied by the fission cross section of the detector (94% U-235). This tally was compared for the 4 different source locations and the FOV was calculated. These results are compared to the PENTRAN results in Table 5. The agreement is good between the two methods.

Table 5: Relative Assembly Importance in PENTRAN and MCNP
(NU Fuel)

Assembly #	PENTRAN FOV	MCNP FOV	MCNP Uncertainty
1	88.55%	87.43%	0.0026%
2	5.35%	6.29%	0.45%
3	5.20%	5.18%	0.59%
4	0.91%	1.10%	6.1%

The field of view of a gamma detector was also investigated. Since there is a wide range of detector possibilities, the one assumed in this paper was uniformly sensitive to all energies. The model used was identical to the neutron detector model except for the adjoint source (uniform). A coupled neutron and gamma calculation was performed, determining the importance of both gammas and neutrons (through n,γ) reactions to the gamma detector. FOV results are shown in Table 6. Different burnups were not tested due to the linear nature of the gamma source with burnup (see Figure 5). In all cases, the response due to (n,γ) reactions was negligible.

Table 6: FOV of Uniform Gamma Detector
(NU Fuel)

Cooling Time (years)	Assembly Number				Fraction contribution from neutrons (n,γ)
	1	2	3	4	
1	88.11%	6.26%	5.06%	0.57%	5.58E-09
5	88.75%	5.97%	4.77%	0.51%	1.83E-08
20	89.02%	5.85%	4.65%	0.48%	3.12E-08

Conclusions

The detector importance calculations we have performed show that the field of view of a detector in the Atucha-I pool extends beyond the nearest 4 assemblies. In a pool of natural uranium fuel, for a fission chamber neutron detector, 88% of the response comes from the nearest 4 assemblies (i.e. one assembly in each direction), while 99% is from the nearest 16 (i.e two assemblies in each direction). A forward MCNP calculation obtained similar results (87% of response from nearest 4 assemblies). For a uniformly sensitive gamma detector, the corresponding results are 89% and 99%, respectively. Gamma response due to (n,γ) reactions are on the order of 10^{-8} and can be neglected. The field of view of both detectors varies very little with burnup and cooling time.

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

References

1. Ham, Y. et al. "Neutron Measurement Techniques for Verification of Closely Packed Spent Fuel Assemblies Stored in a Spent Fuel Pond," Proceedings of the INMM 48th Annual Meeting, Tucson, AZ, July 2007.